INVESTIGATING THE FILLING STATE OF OSL DETECTOR TRAPS
WITH THE OPTICAL SAMPLING METHOD

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Abstract
Optically stimulated luminescence (OSL) and thermoluminescence (TL) methods are commonly used in dosimetry of ionizing radiation and dating of archaeological and geological objects. A typical disadvantage of OSL detectors is signal loss over a longer time scale. In this article, we present a method of studying this phenomenon as well as monitoring the state of the detector by means of optical sampling. The method was used to determine the OSL signal loss (fading) characteristics of selected potassium feldspars.

Keywords: optically stimulated luminescence (OSL), ionizing radiation detectors, radiative recombination, signal loss.

1. Introduction

Ionizing radiation (especially X-rays and gamma rays) has found a wide application in various areas of life, such as medicine (X-ray imaging, tomography, radiotherapy), metal industry (defectoscopy), food preservation and sterilization, archaeological and geological dating, airport security and many more.

In many of these applications the radiation doses are significant (e.g., tomography, 1–60 mGy effective dose per scan [1]) and sometimes very high (e.g., food preservation and sterilization, even several kGy [2]). Therefore, they can potentially threaten human health and life. However, even for small doses, there is a need for constant radiological protection through systematic measurement of individual doses and radiological monitoring of the environment.

Modern ways of detecting ionizing radiation are increasingly based on luminescence methods which allow the determination of the dose of ionizing radiation absorbed by a human or other material object in a fairly simple and accurate way [1, 3–7]. Information about the absorbed dose can be read by subjecting the luminescence detector to appropriate stimulation – thermal...
or optical. The stimulated detector emits a weak light (luminescence), the amount of which is proportional to the absorbed dose of radiation. The luminescence produced by heating is called thermoluminescence (TL). TL detectors have been used in ionizing radiation dosimetry for several dozen years [4–6]. However, precise temperature control (heating) is quite difficult. The TL reader is therefore a complex and expensive device [7].

Another method of extracting information on absorbed dose from an irradiated detector is the optically stimulated luminescence (OSL) method. Stimulation is most often performed with monochromatic light with constant intensity (CW-OSL), linearly modulated optically stimulated luminescence (LM-OSL) or pulsed light optically stimulated luminescence (POSL) [1, 8]. The main idea of the OSL method is the registration of emission in the range of wavelengths shorter than those of the stimulating light. This distinguishes this method from the better-known photoluminescence [9].

The TL or OSL detector is a passive element that does not require any electronics or power supply. Usually, it is a solid dielectric with a crystal or polycrystalline structure. These detectors, using long-life luminescence, have the feature of remembering and collecting information about the absorbed dose of ionizing radiation for a very long time – even thousands of years [10].

Optical stimulation is easier to implement than thermal stimulation [11]. Unfortunately, the amount of materials suitable for OSL dosimetry is much smaller than for TL dosimetry. The basic OSL material is the Al₂O₃:Cr crystal which was introduced for commercial use by Landauer (USA) in the 90s. The irradiated Al₂O₃:Cr detector emits luminescence in the UV range when stimulated with visible light (e.g., blue or green) [1, 8].

Another commercially available OSL detector is BeO [12]. The material offers an alternative to Al₂O₃:Cr in OSL dosimetry. BeO crystals exhibit high sensitivity to ionizing radiation and a linear dose response over six orders of magnitude. It is also nearly tissue-equivalent. Nevertheless, its potential use in routine dosimetry is somewhat limited due to high toxicity of this material, especially in the powder form [13–15].

The role of a luminescent detector can also be played by materials commonly found in the human environment – such as minerals (e.g., quartz, feldspar, rock salt) or some items of common use (e.g., porcelain, bricks, electronic components, table salt, etc.). These materials play a large role in geological and archaeological dating and the so-called emergency dosimetry. In the latter case, it is necessary to use a “replacement detector” when a radiation event has occurred and there were no devices on site that could register this radiation [16–18].

In recent years, a lot of research has been carried out in order to find new materials that can act as luminescent detectors – both new ones, synthesized in laboratories for the purposes of routine dosimetry, and those that occur in nature or can be found in objects of common use. This applies in particular to materials that can be used in OSL dosimetry [19].

Unfortunately, many suitable materials, even those that exhibit strong luminescence, also suffer from some disadvantages of loss of TL or OSL signal during long storage. The phenomenon of signal loss (fading) is quite common [17, 20, 21]. Nevertheless, it does not completely rule out the suitability of the material as a TL or OSL detector. However, an appropriate correction should be taken into account and/or the detector should be subjected to a special measurement procedure.

Various measurement methods are used to investigate the loss of the signal. They usually consist in making a large series (several dozen) of samples and testing the TL or OSL response after a sufficiently long period of time. Such a test requires high homogeneity of the measured samples and is very time-consuming.

In this article, we present a method of examining the state of an OSL detector using optical sampling. The method does not require so many (several dozen) samples, so it is much faster. To determine a fading curve only two pulsed OSL measurements are required.
strate its usefulness, this method was applied to the microcline mineral which belongs to the group of potassium feldspars. The minerals are natural OSL detectors often used in geological dating [22, 23].

2. Theoretical basics

Let us consider the most common case where the OSL detector is a dielectric with a crystalline structure. The energy gap in such material is quite large i.e. above 5 eV [1, 5]. The existence of long-lived luminescence, the lifetime of which is counted in years, is possible due to structural defects. These defects may correspond to typical imperfections of the crystal lattice (vacancies, interstitial positions) as well as impurities introduced spontaneously or intentionally into the material. Defects create localized energy levels within a wide band gap. These levels act as traps and recombination centres for the majority of charge carriers. A simplified model of the irradiation stage and the OSL emission is shown in Fig. 1. It assumed that only electrons occupy “active” traps and may be released to the transport band by OSL stimulation.

![Diagram of OSL detector](image)

Fig. 1. Simplified diagrams of localized energy levels and charge carrier transitions in an OSL detector. The upper diagram presents charge carrier transitions during the irradiation stage. Ionizing radiation (X) produces free electrons and holes which may be trapped (A_i, A_M,l and A’k) or recombine (B_k). The bottom diagram presents charge carrier transitions during OSL measurement. Charge carriers (here electrons) released during OSL stimulation (D_i) may recombine with trapped holes (B_k) producing radiative OSL emission or be retrapped (A_i). The T transition represents direct recombination which may cause an OSL signal decay.
During excitation (irradiation), the localized energy levels (traps and recombination centres) are filled with electrons and holes, respectively. Luminescence can occur through direct recombination between electrons in traps and holes in recombination centres (the so-called localized transitions model, LT) [24,25]. Recombination is also possible through delocalized bands (STM – the simple trap model) [26]. The latter case is more often considered.

Many of the trap levels are so deep that electrons remain permanently in them (with a lifetime of more than thousands of years) and no radiative recombination (and therefore luminescence) occurs. To speed it up, we use detector stimulation. Heating to a sufficiently high temperature (200–300°C) significantly increases the probability of releasing electrons from the traps and enables their recombination. This case refers to the phenomenon of thermoluminescence (TL).

Another way to free electrons from traps is by light stimulation. Typically, this is strong monochrome light in the visible range (VIS) or near infrared (IR). In the OSL reader, optical detection filters practically cut-off the stimulation component, registering the detector’s weak emission in the range of shorter wavelengths – usually UV [1,8,27,28].

Nevertheless, many TL and OSL detectors and natural materials do not follow this simple scheme. Despite having deep traps, these materials lose their charge carriers over time. The mechanism of this phenomenon may be different. Often there is a non-radiative (or radiative, but beyond the range of observation) recombination bypassing the conduction band. This may be the case when traps and recombination centres are close together. Then recombination may result from tunnelling (cf. transition T in Fig. 1) [18].

In this case, the loss in the concentration of electrons (fading) at the active trap levels $n$ can be represented by the equation:

$$\frac{dn(t)}{dt} = -\alpha n,$$

which leads to a simple exponential decay:

$$n(t) = n_0 e^{-\alpha t} = n_0 e^{-t/\tau}.$$

In the equations above $n_0$ denotes the initial concentration of electrons at time $t = 0$, $\alpha$ is the recombination rate which is typically presented in the form of a characteristic decay time, i.e., $\tau = \alpha^{-1}$.

The relation above is consistent with many experimental observations, but such a relationship is not a rule and can sometimes have a more complex form [17,18]. In the present considerations, however, we will adopt the simplest formula (2).

3. OSL sampling – theory

The readout of a TL detector frees all charge carriers from active traps. The heating resets the detector state and no further reading is possible. The OSL method has an advantage in this respect [1,8]. Using a suitably short light pulse, only some of the carriers can be released from traps. In this way, we obtain information about the current state of the detector (i.e., the density of occupied trap levels) and this measurement can be repeated many times. This can be done in most cases, except extremely low-dose measurements, where the whole population of trapped charge carriers must be used to detect a small OSL signal.

It seems that the possibility of sampling the state of an OSL detector allows also to determine the fading constant $\tau$. However, there is a problem – each measurement (the short light pulse) decreases the number charge carriers in active traps and can significantly change (decrease) the value of the calculated fading constant. Let us analyse this situation quantitatively.
By illuminating the detector with a beam of light $\Phi$ during the time $\tau_s$, we cause a loss of electrons at the trap levels in the amount:

$$\Delta n_1 = -\chi \Phi \tau_s n(t_1) = -\chi \Phi \tau_s n_0 e^{-t_1/\tau}. \quad (3)$$

In the formula above, we assume that the measurement was made at time $t_1$ when the concentration of carriers in the traps was $n_1 = n(t_1)$. Formula (3) is correct in the case of very short pulses when the number of excited carriers is significantly lower than the total maximum occupancy of $N$ levels $i.e.$, $|\Delta n_1| \ll N$. The constant $\chi$ determines the efficiency of OSL stimulation. In the case of a significant loss of charge carriers, with a small number of recombination centers, the value of $\chi$ may change, but we will not consider this situation here.

After illuminating the detector with a single pulse, the number of carriers in active traps will decrease to the following level:

$$\tilde{n}_1(t_1) = n_0 e^{-t_1/\tau} - \chi \Phi \tau_s n_0 e^{-t_1/\tau} = n_0 e^{-t_1/\tau} \left(1 - \chi \Phi \tau_s \right). \quad (4)$$

Similarly, after the $k$-th pulse, the occupation of the trap levels will be:

$$\tilde{n}_k(t_k) = n_0 e^{-t_k/\tau} \left(1 - \chi \Phi \tau_s \right)^k. \quad (5)$$

It is easy to see that in the absence of external influence ($i.e.$, detector sampling), the filling of trap levels would have the value of:

$$n_k(t_k) = n_0 e^{-t_k/\tau}. \quad (6)$$

These values can be “recovered” by appropriately normalizing carrier concentrations resulting from the detector sampling procedure as follows

$$n_{k,\text{norm}}(t_k) = \tilde{n}_k(t_k) \left(1 - \chi \Phi \tau_s \right)^{-k}. \quad (7)$$

For this purpose, it is necessary to calculate the factor $\left(1 - \chi \Phi \tau_s \right)$ which, for the sake of simplicity, we denote here by $\xi$. Of course, in a real OSL measurement, we do not directly measure the filling of the active trap levels. Nevertheless, under the conditions described previously, $i.e.$, short pulse stimulation and a relatively small loss of carriers in the recombination centers, we can assume that the total luminescence recorded during and immediately after the end of the stimulation pulse is proportional to the current concentration of carriers (electrons) at the active trap levels $n(t)$:

$$\mathcal{L}(t) = \gamma \Phi \tau_s n(t). \quad (8)$$

The equation can be explained as follows: according to the model presented in Fig. 1, luminescence arises as a result of recombination of free electrons with trapped holes. Thus, the luminescence has to be proportional to the number of charge carriers released to the conduction band by a short light pulse. This, in turn, is proportional to the actual concentration of carriers in the traps $n(t)$. The constant $\gamma$ determines the luminescence efficiency during the pulsed OSL stimulation. Its nature and limitations are similar to the previously defined constant $\chi$ (3). Thus, we will continue to assume that its value does not change either.

Now we will show how by making a series of pulse OSL measurements over a short period of time $\Delta t_m$ – significantly shorter than the characteristic decay (fading) time, $i.e.$, $\Delta t_m \ll \tau$, the factor $\xi$ can be determined. Under these conditions, we can neglect the exponential factor
that appears in (2) and (5). Subsequent luminescence measurements will then give the following results:

\[
\begin{align*}
L_0 &= \gamma \Phi \tau_s n_0 \\
L_1 &= \gamma \Phi \tau_s n_0 \xi \\
\vdots \\
L_k &= \gamma \Phi \tau_s n_0 \xi^k
\end{align*}
\] (9)

Taking the logarithm of the last dependence, we get the equation:

\[
\ln(L_k) = C + k \ln \xi,
\] (10)

which in the graph \(\ln(L_k) \times k\) shows a straight line with a slope \(\ln \xi\). In this equation, \(C\) is a constant.

The factor \(\xi\) indicates the relative number of electrons that will remain in the active traps after the end of a single pulse stimulation. Our knowledge of it allows to normalize the luminescence values obtained in the OSL detector sampling test over a long time scale. Thanks to this, it is possible to determine the characteristic decay time of the signal \(\tau\) of a given detector by calculating the corrected values of luminescence according to the following formula:

\[
L_{k,\text{norm}} = \tilde{L}_k \xi^{-k},
\] (11)

where \(\tilde{L}_k\) denotes the luminescence signal (e.g., the number of counts) measured during (and shortly after) the \(k\)-th OSL pulse. Then, using the relationship determined directly by (2) and (8) we have

\[
L_{k,\text{norm}}(t) = L_0 e^{-t/\tau}.
\] (12)

4. Materials and methods

Potassium feldspars are common minerals. For years, they have aroused the interest of geologists due to their ability to collect information about the absorbed dose of ionizing radiation over a long period of time [22,23]. Thus, feldspars are natural detectors of ionizing radiation. They have the potential to date various geological processes over thousands of years. However, these materials have a very complex structure. They contain many localized energy levels of varying depth and spatial arrangement. Therefore, obtaining the desired dosimetric information is a very difficult task.

Microcline crystals from the Strzegom Massif were used in this research. The samples were irradiated with a \(^{90}\text{Sr}/^{90}\text{Y}\) source of beta radiation with an activity of 2.9 GBq. The irradiation process was carried out in the dark. The dose of 60 Gy was used. The minerals exhibit significant fading in the range of several hours/days. The dose 60 Gy was high enough to observe the fading for a sufficiently long period of time. The sample was a single grain crystal. Prior to each irradiation the sample was optically bleached for 30 min using IR diodes of the OSL reader.

OSL readings were made using a HELIOS-3 OSL reader [27,28]. For feldspar measurements, the reader was equipped with 15 VSLY5850 IR diodes with a radiant power of 55 mW each which stimulated the samples in the range of 840–870 nm (peak 850 nm). The light emitted by the LEDs was additionally filtered with 3 mm Schott RG715 filters.

Luminescence detection was performed in the UV-VIS 300–600 nm range with a maximum at approx. 500 nm, using 2.5 mm Schott BG39 filters placed in front of an integrated H7360
(Hamamatsu) photon counting module with a quartz window. The data were collected by a dedicated electronic system which sent them to a computer. The Helios reader is also equipped with an additional photosensor used to control the emission of LEDs.

5. OSL sampling – results for a microcline crystal

The OSL sampling for a single grain microcline crystal (22.4 mg) was performed using short IR light pulses lasting $\tau_s = 1$ s. The stimulation pulses were applied every 300 s and the OSL signal was recorded every 0.1 s. The result in the range of approx. 100 minutes is shown in Fig. 2. Control measurements, made to calculate the $\xi$ coefficient, were made in the same conditions, but for a shorter time scale (Fig. 3). In this measurement the same sample was used after IR bleaching for 30 min. The stimulation pulses were applied every 11 s. This time is sufficient for the decaying luminescence to reach the background. The integrated luminescence signal for each pulse was plotted (Fig. 4) against the measurement number ($k$) according to (10). The integration was performed in the range of 5 s. Fitting the straight line to this plot gave the result of the normalization factor $\xi = 0.98515 \pm 0.00013$.

Fig. 2. Result of OSL sampling of a natural detector made of a microcline mineral.

Fig. 3. Result of OSL control measurements of the natural detector made of microcline carried out in a shorter time scale to normalize the sampling measurements (Fig. 2) according to (11).
The next step is to calculate the corrected values of luminescence of the material during fading. For this purpose, the integrated luminescence signal for each pulse of the measurement shown in Fig. 2 was calculated and multiplied by $\xi^{-k}$ according to (11). The result is shown in Fig. 5.

Further calculations with the use of (11) and (12) allow to estimate the characteristic fading time $\tau$ of the tested emission component at approx. $5.0 \times 10^4$ [s], i.e., approx. 14 hours ($\tau = 13.9 \pm 0.8$ h). The result agrees well with the value of the fading time obtained with a more typical traditional method.

6. Conclusions

The article presents the method of studying OSL signal loss (fading) in various detectors of ionizing radiation. The method consists in optical “probing” of the detector state by means of pulsed OSL stimulation. The resulting luminescent signal must be corrected. For this purpose, we perform a second measurement – on a much shorter time scale, which allows to calculate the normalization factor and restore the correct characteristics of the signal loss of the detector.
The advantage of the proposed method is simplicity with a smaller number of samples and much shorter time required for measurements. Typical determination of a fading curve is very time-consuming. It needs several dozen of OSL measurements that require preparation of several dozen samples and the same number of irradiations. Additional procedures involve also administration of calibration or test doses to the samples [29]. The proposed method needs only two samples (or one with bleaching, as implemented by us) and two pulsed OSL measurements. Hence, it is much faster.

The idea of the method was presented in application of a microcline crystal from the Strzegom Massif as a natural detector of ionizing radiation. The characteristic fading time of this mineral was estimated to approx. 14 hours.

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References


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